SUPPLEMENTARY MATERIAL FOR

Hyperfine Coupling and Slow Magnetic Relaxation in Isotopically Enriched Dy^{III} Mononuclear Single-Molecule Magnets

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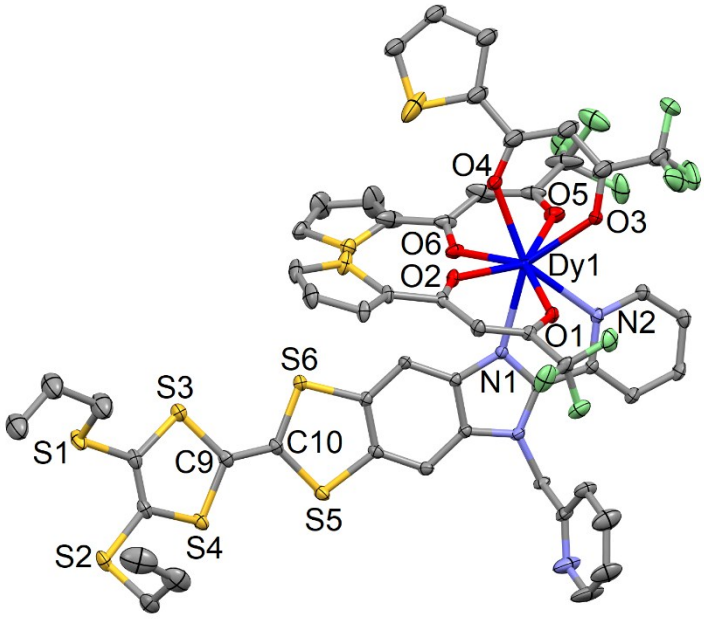

Figure S1. ORTEP view of the asymmetric unit in **^{162}Dy**. Thermal ellipsoids are drawn at 30% probability. Hydrogen atoms and *n*-hexane molecule of crystallization are omitted for clarity.

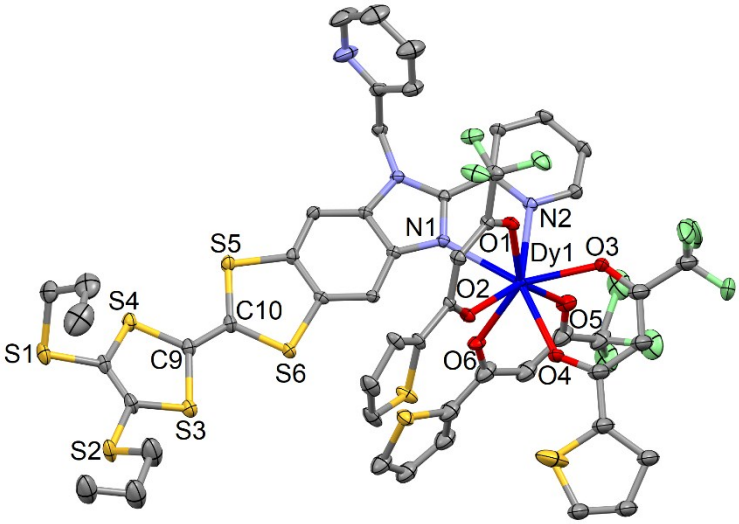

Figure S2. ORTEP view of the asymmetric unit in **^{163}Dy**. Thermal ellipsoids are drawn at 30% probability. Hydrogen atoms and *n*-hexane molecule of crystallization are omitted for clarity.

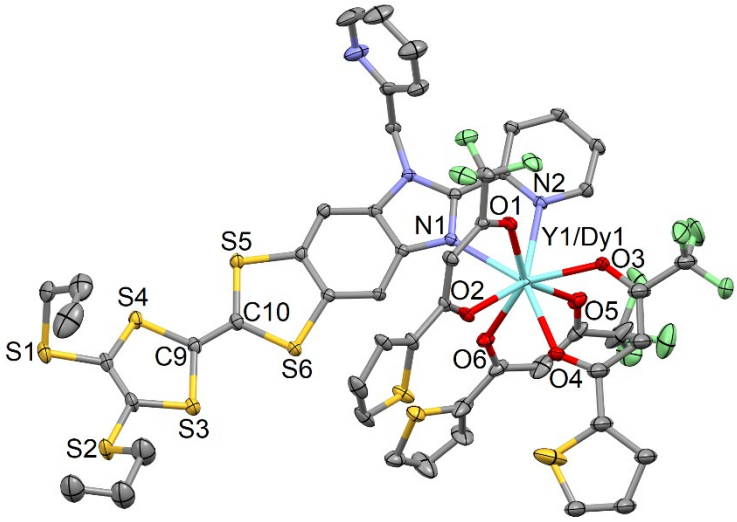

Figure S3. ORTEP view of the asymmetric unit in **^{162}Dy@Y**. Thermal ellipsoids are drawn at 30% probability. Hydrogen atoms and *n*-hexane molecule of crystallization are omitted for clarity.

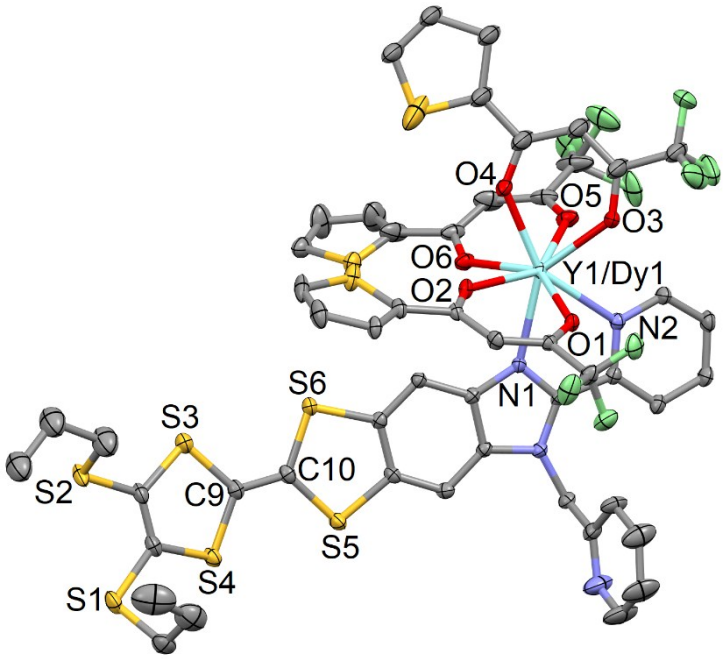

Figure S4. ORTEP view of the asymmetric unit in **^{163}Dy@Y**. Thermal ellipsoids are drawn at 30% probability. Hydrogen atoms and *n*-hexane molecule of crystallization are omitted for clarity.

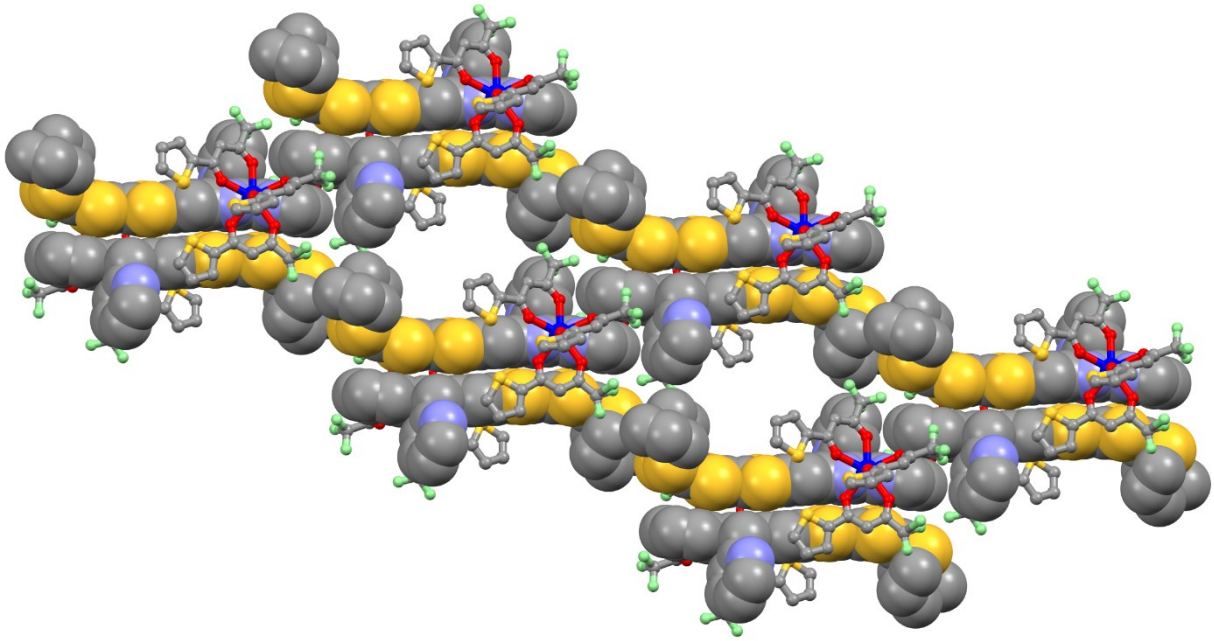

Figure S5. Crystal packing view of **^{162}Dy** highlighting the formation of "head-to-tail" dimers. The ligand **L** and Dy(tta)$_3$ organometallic are respectively shown in "space fill" and "ball and sticks" representations.

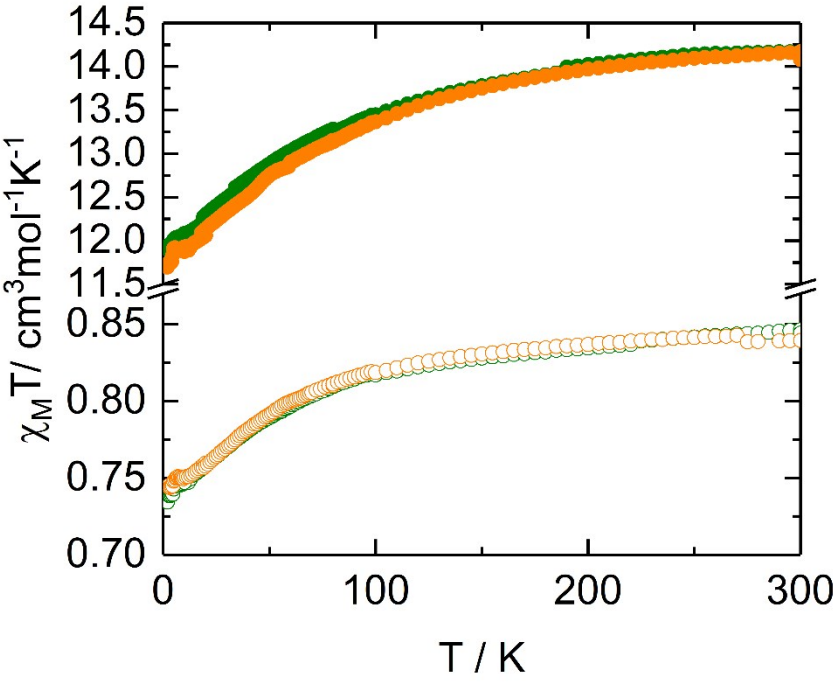

Figure S6. Thermal variations of $\chi_M T$ of a solid-state samples of **^{162}Dy** (full orange circles), **^{163}Dy** (full green circles), **^{162}Dy@Y** (open orange circles) and **^{163}Dy@Y** (open green circles) between 2 and 300 K in applied magnetic field of 200 Oe for temperatures between 2 and 20 K, 2 kOe between 20 and 80 K and 10 kOe above.

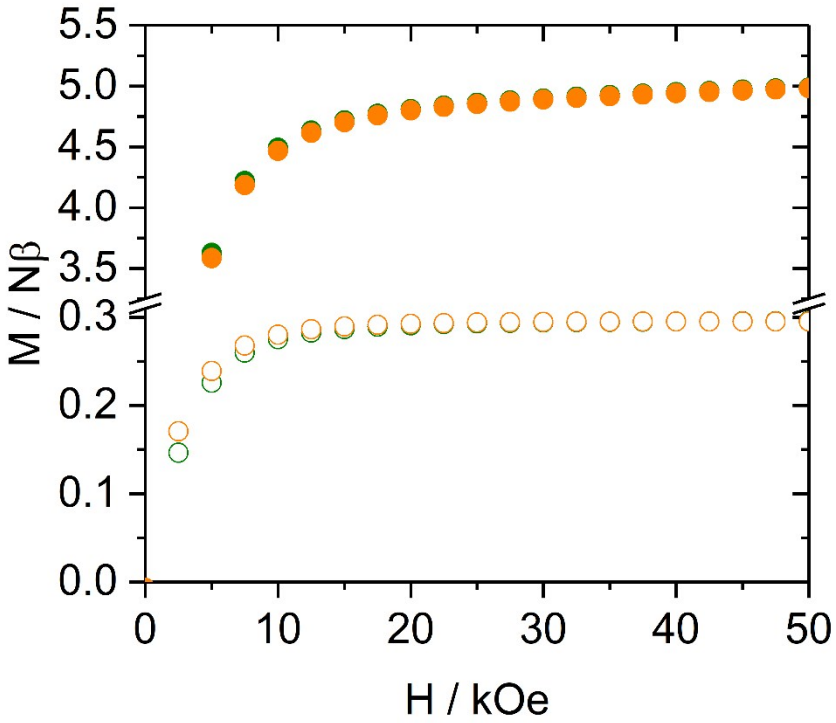

Figure S7. Field variations of the magnetization at 2 K for **^{162}Dy** (full orange circles), **^{163}Dy** (full green circles), **^{162}Dy@Y** (open orange circles) and **^{163}Dy@Y** (open green circles).

Extended Debye model.

$$\chi_M' = \chi_S + (\chi_T - \chi_S)\frac{1+(\omega\tau)^{1-\alpha}\sin\left(\alpha\frac{\pi}{2}\right)}{1+2(\omega\tau)^{1-\alpha}\sin\left(\alpha\frac{\pi}{2}\right)+(\omega\tau)^{2-2\alpha}}$$

$$\chi_M'' = (\chi_T - \chi_S)\frac{(\omega\tau)^{1-\alpha}\cos\left(\alpha\frac{\pi}{2}\right)}{1+2(\omega\tau)^{1-\alpha}\sin\left(\alpha\frac{\pi}{2}\right)+(\omega\tau)^{2-2\alpha}}$$

With χ_T the isothermal susceptibility, χ_S the adiabatic susceptibility, τ the relaxation time and α an empiric parameter which describe the distribution of the relaxation time. For SMM with only one relaxing object α is close to zero. The extended Debye model was applied to fit simultaneously the experimental variations of χ_M' and χ_M'' with the frequency ν of the oscillating field ($\omega = 2\pi\nu$). Typically, only the temperatures for which a maximum on the χ_M'' vs. ν curves, have been considered (see figure here below for an example). The best fitted parameters τ, α, χ_T, χ_S are listed in Table S2 to S7 with the coefficient of determination R^2.

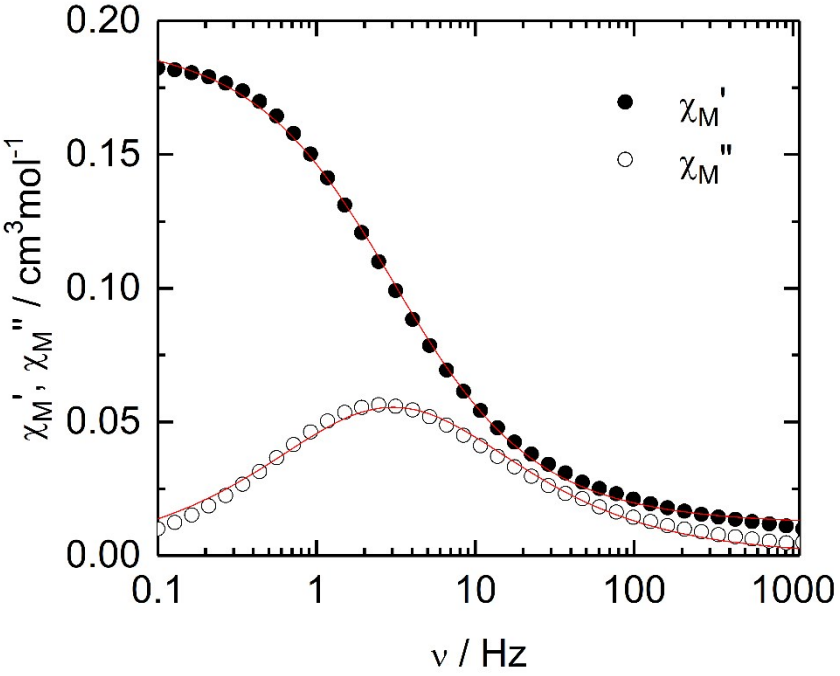

Figure S8. Frequency dependence of the in-phase (χ_M') and out-of-phase (χ_M'') components of the ac susceptibility measured on powder at 4 K in zero dc field with the best fitted curves (red lines) for **^{163}Dy@Y**.

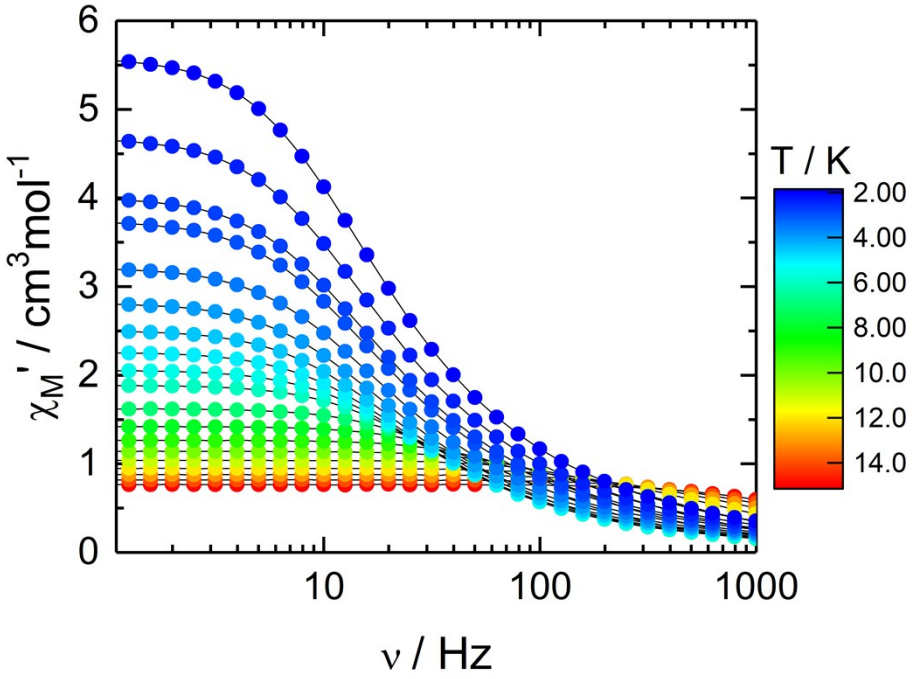

Figure S9. Frequency dependence of the in-phase component of the ac susceptibility measured in zero dc field for compound **^{162}Dy** between 2 and 15 K.

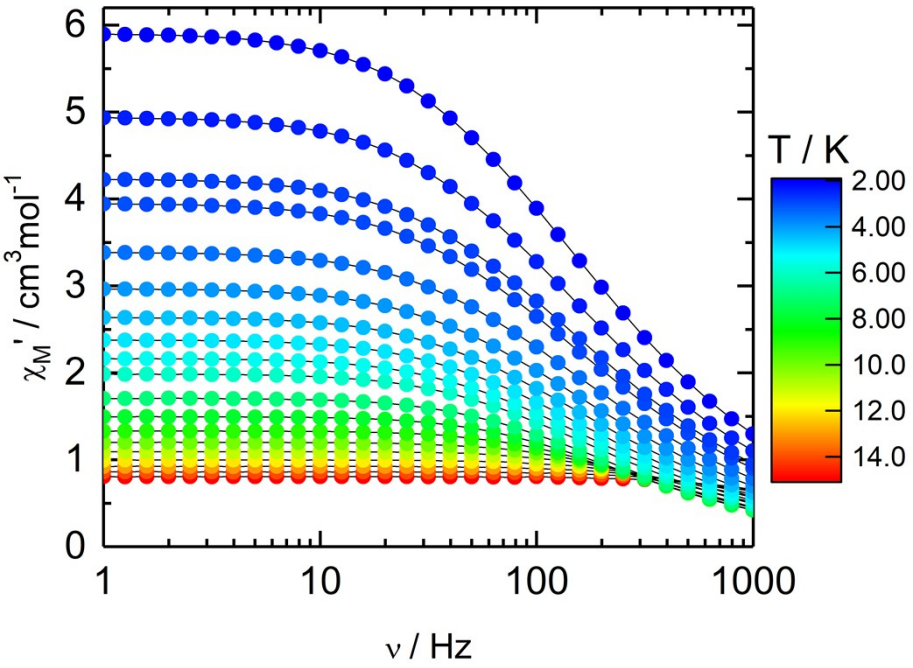

Figure S10. Frequency dependence of the in-phase component of the ac susceptibility measured in zero dc field for compound **^{163}Dy** between 2 and 15 K.

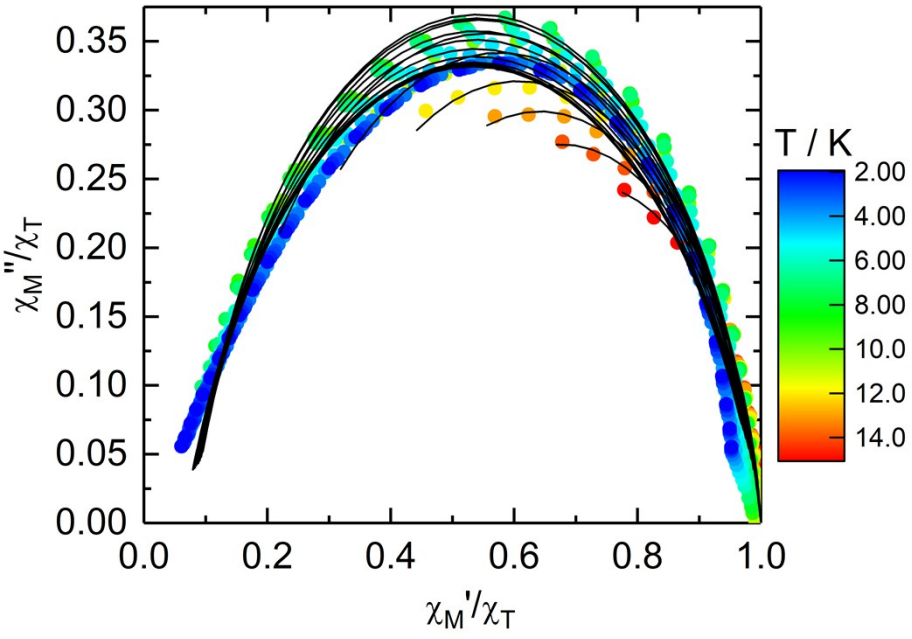

Figure S11. Normalized Cole-Cole plots in zero magnetic field for **^{162}Dy** between 2 and 15 K.

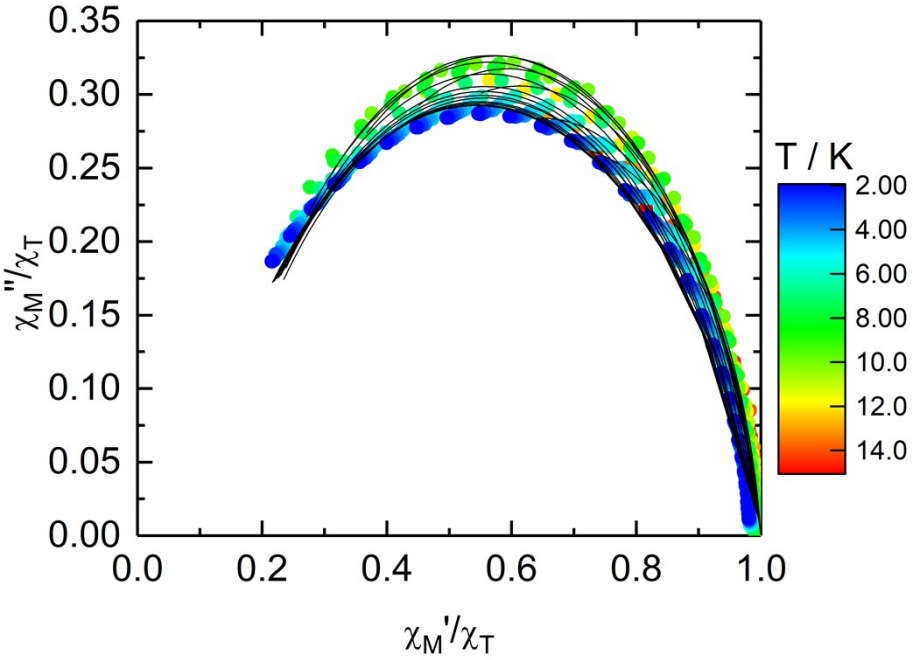

Figure S12. Normalized Cole-Cole plots in zero magnetic field for **^{163}Dy** between 2 and 15 K.

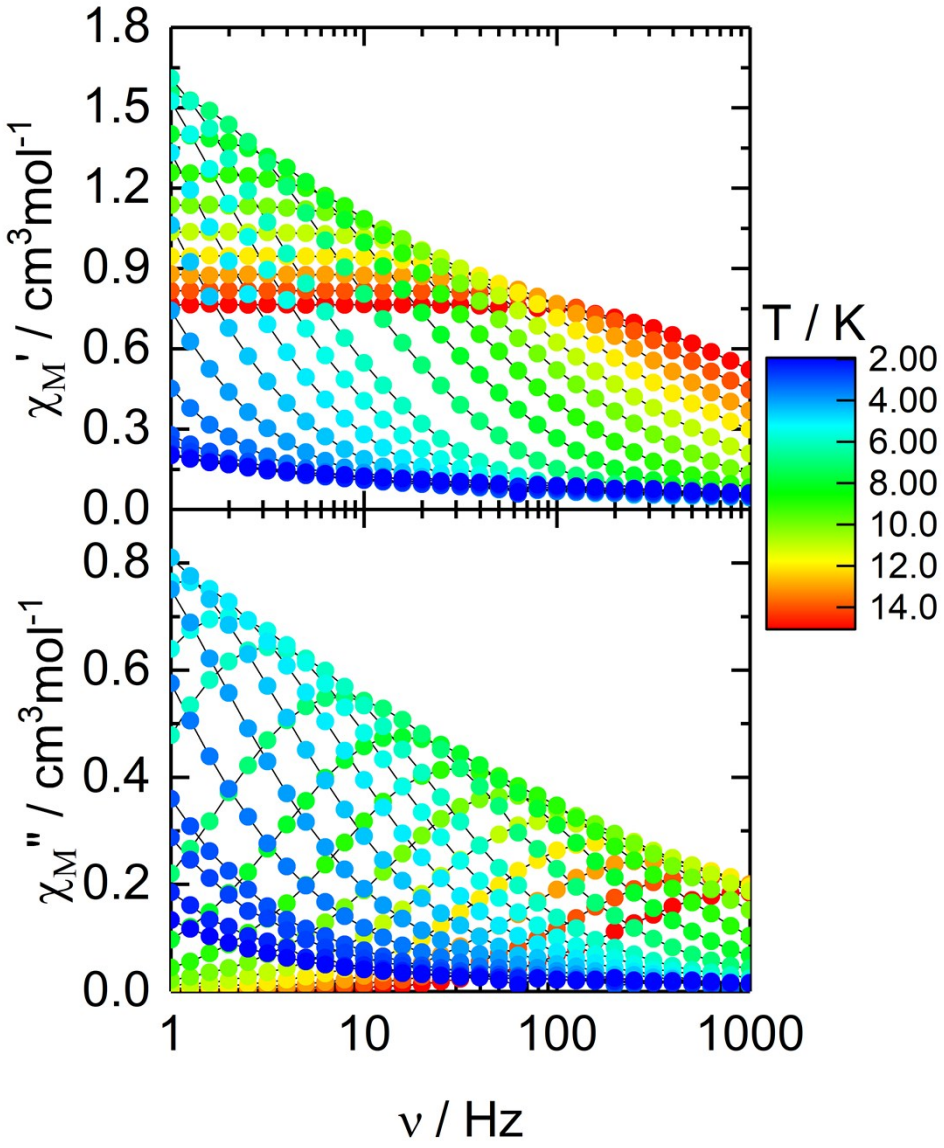

Figure S13. Frequency dependence of the in-phase (top) and out-of-phase (bottom) components of the ac susceptibility measured in 1000 Oe dc field for compound **^{162}Dy** between 2 and 15 K.

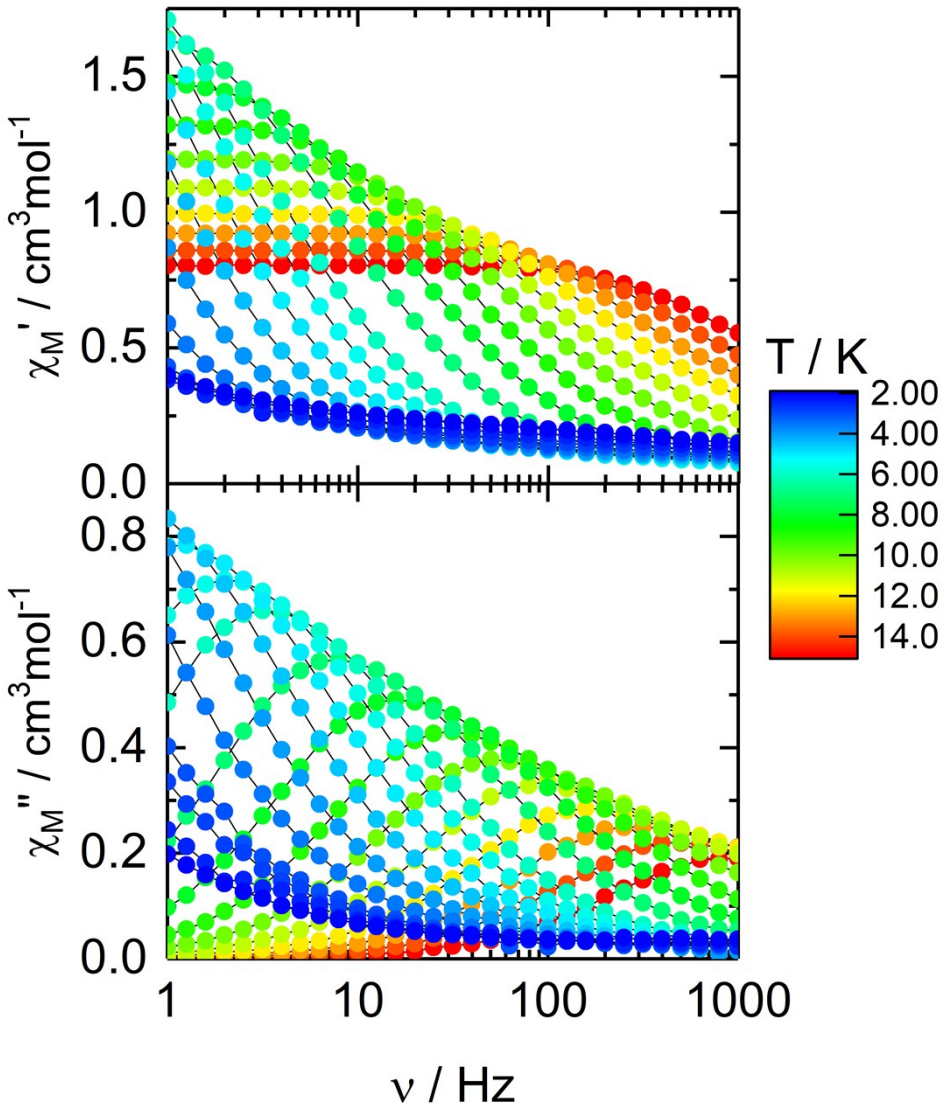

Figure S14. Frequency dependence of the in-phase (top) and out-of-phase (bottom) components of the ac susceptibility measured in 1000 Oe dc field for compound **^{163}Dy** between 2 and 15 K.

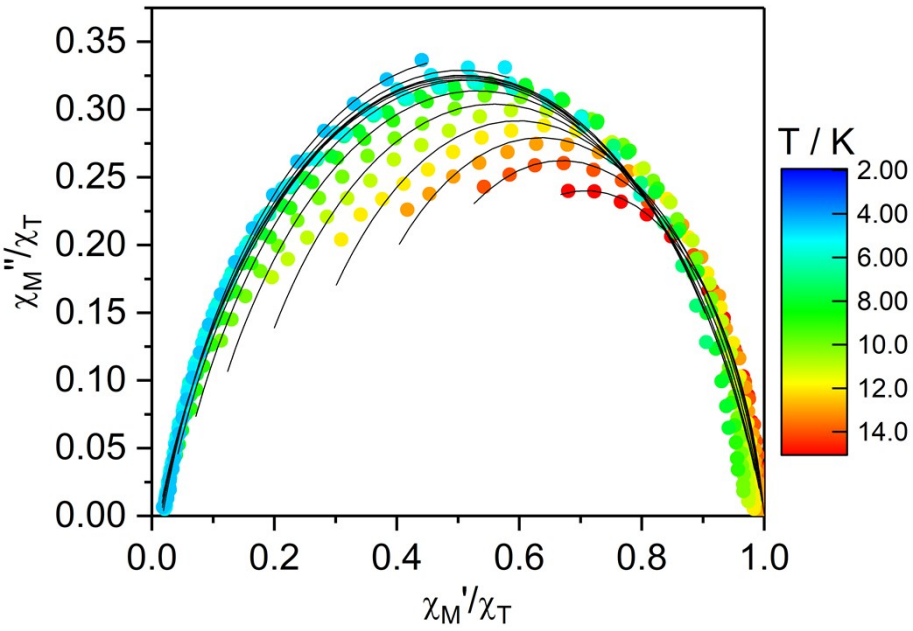

Figure S15. Normalized Cole-Cole plots in 1000 Oe magnetic field for **^{162}Dy** between 2 and 15 K.

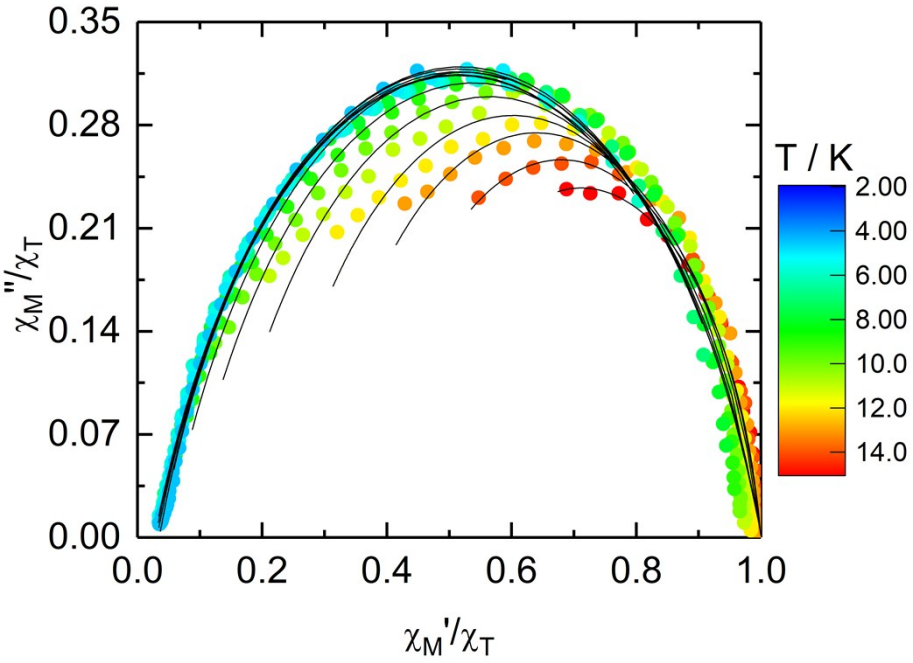

Figure S16. Normalized Cole-Cole plots in 1000 Oe magnetic field for **^{163}Dy** between 2 and 15 K.

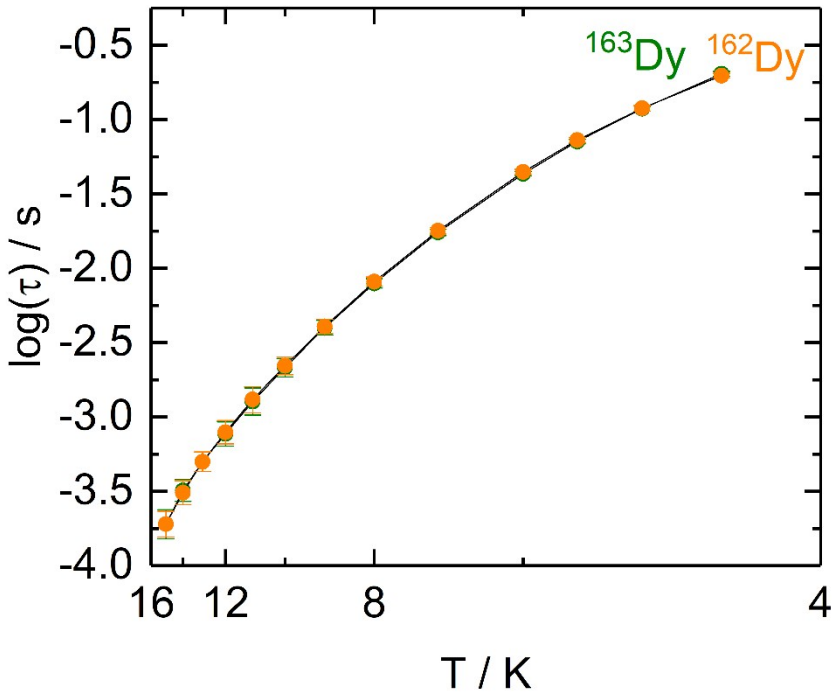

Figure S17. Log scale plots of the temperature dependence of the relaxation time of **^{162}Dy** (full orange circles) and **^{163}Dy** (full green circles) measured in 1000 Oe magnetic field.

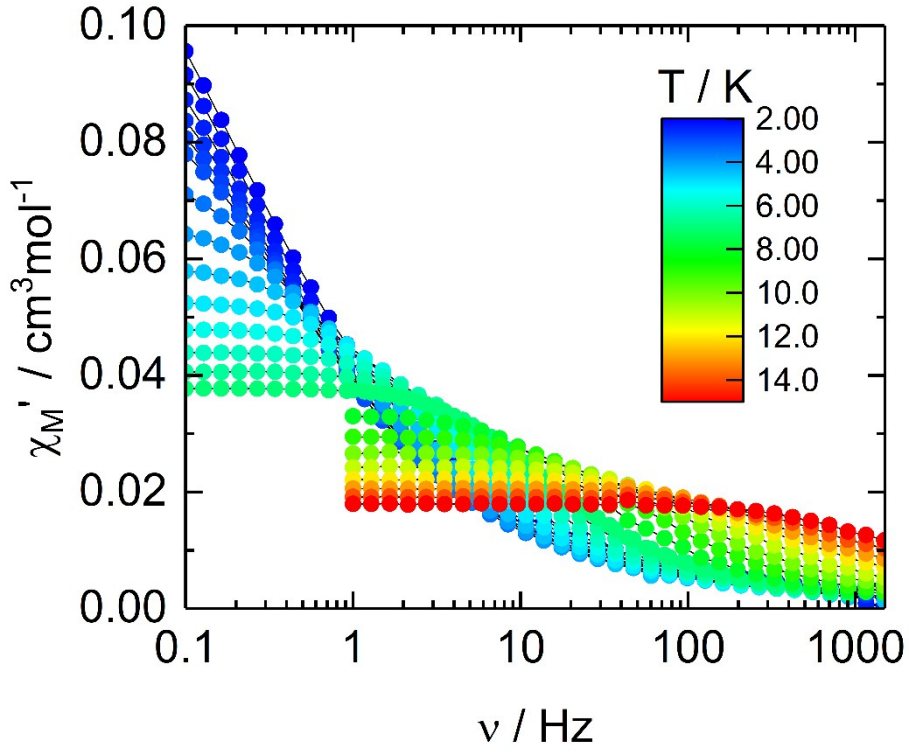

Figure S18. Frequency dependence of the in-phase component of the ac susceptibility measured in zero dc field for compound **^{161}Dy@Y** between 2 and 15 K.

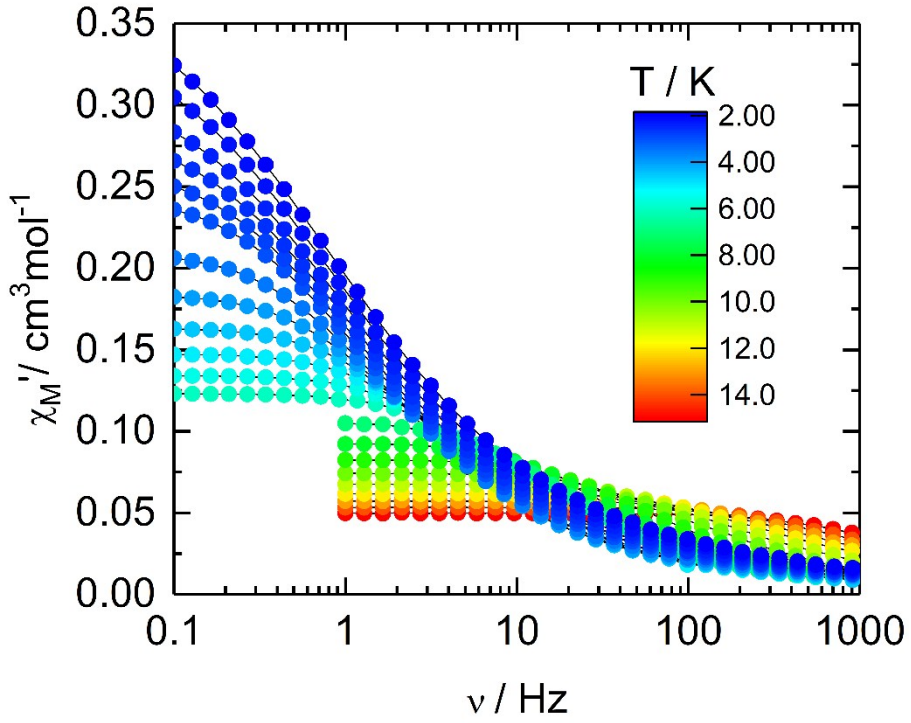

Figure S19. Frequency dependence of the in-phase component of the ac susceptibility measured in zero dc field for compound **^{163}Dy@Y** between 2 and 15 K.

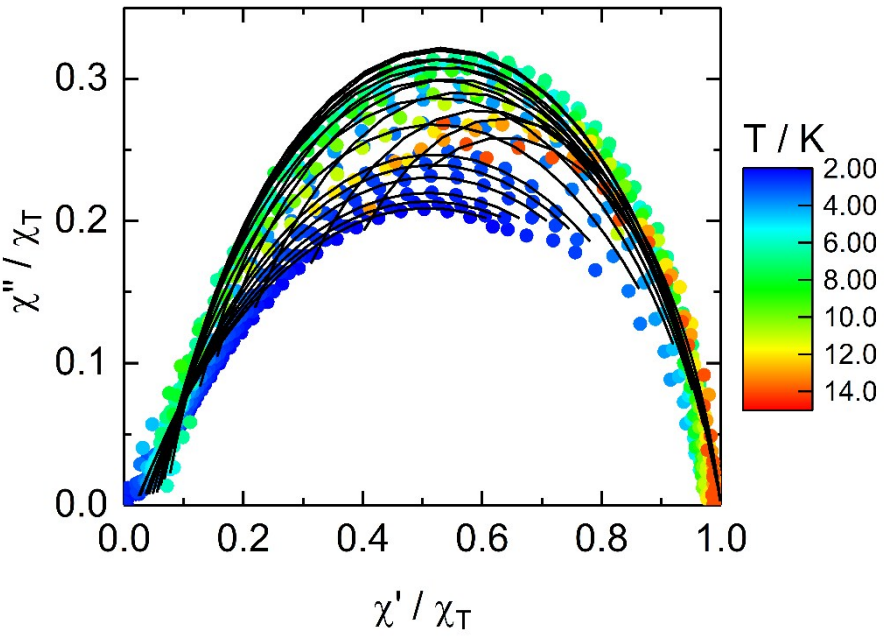

Figure S20. Normalized Cole-Cole plots in zero dc field for **^{161}Dy@Y** between 2 and 15 K.

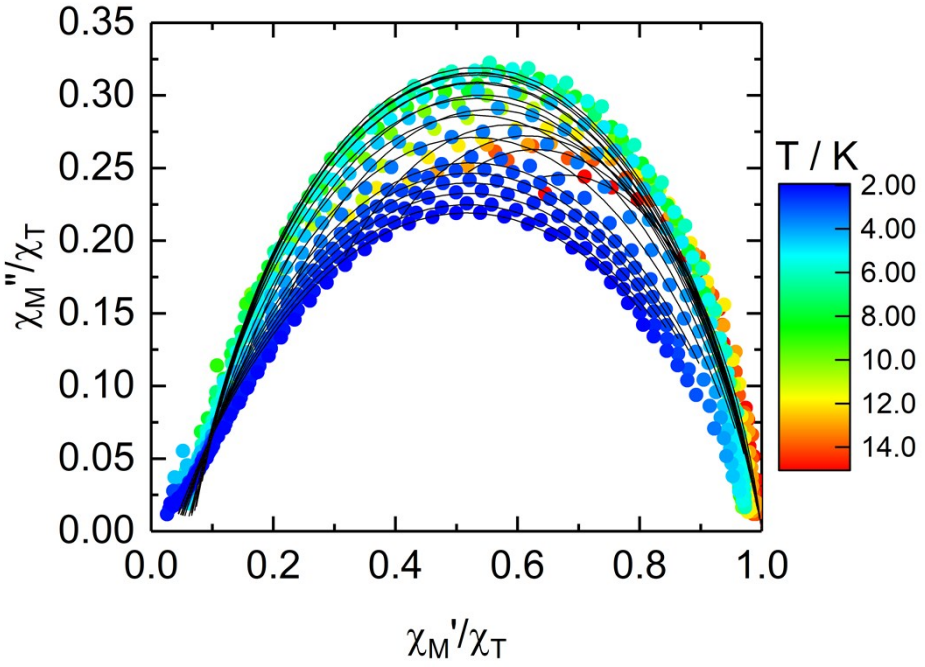

Figure S21. Normalized Cole-Cole plots in zero dc field for **^{163}Dy@Y** between 2 and 15 K.

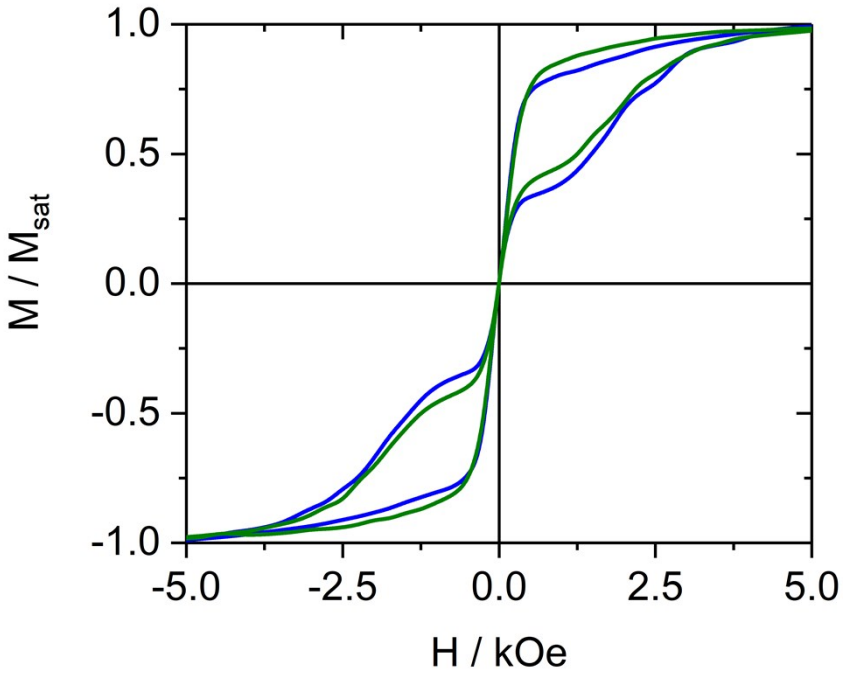

Figure S22. Magnetic hysteresis loops measured on **^{161}Dy@Y** and **^{163}Dy** at 0.46 K at a sweep rate of 16 Oe s^{-1}.

Table S1. X-ray crystallographic data for the complexes **^{162}Dy**, **^{162}Dy@Y**, **^{163}Dy** and **^{163}Dy@Y**.

Compounds	$[^{162}Dy(tta)_3(\mathbf{L})]\cdot(C_6H_{14})$ (**^{162}Dy**)	$[^{162}Dy_{0.05}Y_{0.95}(tta)_3(\mathbf{L})]\cdot(C_6H_{14})$ (**^{162}Dy@Y**)
Formula	$C_{58}H_{52}DyF_9N_4O_6S_9$	$C_{58}H_{52}Dy_{0.05}Y_{0.95}F_9N_4O_6S_9$
M / g.mol^{-1}	1522.57	1452.19
Crystal system	triclinic	monoclinic
Space group	P-1 (N°2)	P-1 (N°2)
Cell parameters	a = 15.3700(12) Å b = 15.7135(13) Å c = 16.7271(14) Å α = 97.981(3)° β = 110.300(3)° γ = 117.242(3)°	a = 15.3310(11) Å b = 15.6911(12) Å c = 16.7368(13) Å α = 98.060(3)° β = 110.194(2)° γ = 117.203(3)°
Volume / Å^3	3146.7(5)	3139.3(4)
Cell formula units	2	2
T / K	150 (2)	150(2)
Diffraction reflection	$5.86 \leq 2\theta \leq 54.96$	$5.86 \leq 2\theta \leq 54.97$
ρ_{calc}, g.cm^{-3}	1.607	1.533
μ, mm^{-1}	1.565	1.308
Number of reflections	67713	68306
Independent reflections	14397	14375
$Fo^2 > 2\sigma(Fo)^2$	12668	11375
Number of variables	783	771
R_{int}, R_1, wR_2	0.0438, 0.0547, 0.1356	0.0527, 0.0731, 0.1962
Compounds	$[^{163}Dy(tta)_3(\mathbf{L})]\cdot(C_6H_{14})$ (**^{163}Dy**)	$[^{163}Dy_{0.05}Y_{0.95}(tta)_3(\mathbf{L})]\cdot(C_6H_{14})$ (**^{163}Dy@Y**)
Formula	$C_{58}H_{52}DyF_9N_4O_6S_9$	$C_{58}H_{52}Dy_{0.05}Y_{0.95}F_9N_4O_6S_9$
M / g.mol^{-1}	1523.57	1453.19
Crystal system	triclinic	triclinic
Space group	P-1 (N°2)	P-1 (N°2)
Cell parameters	a = 15.3548(22) Å b = 15.7144(23) Å c = 16.7281(23) Å α = 98.032(5)° β = 110.191(5)° γ = 117.221(5)°	a = 15.3157(10) Å b = 15.6570(10) Å c = 16.7142(10) Å α = 98.058(2)° β = 110.199(2)° γ = 117.307(2)°
Volume / Å^3	3147.3(8)	3120.8(3)
Cell formula units	2	2
T / K	150 (2)	150(2)
Diffraction reflection	$4.46 \leq 2\theta \leq 55.03$	$5.88 \leq 2\theta \leq 54.97$
ρ_{calc}, g.cm^{-3}	1.607	1.543
μ, mm^{-1}	1.565	1.328
Number of reflections	66322	77501
Independent reflections	14444	14299
$Fo^2 > 2\sigma(Fo)^2$	12519	9999
Number of variables	699	783
R_{int}, R_1, wR_2	0.0670, 0.0606, 0.1515	0.0617, 0.0724, 0.1870

Table S2. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **^{162}Dy** at 0 Oe in the temperature range 2-15 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R²
15	0.76651	0.34054	0.07877	0.00013	0.99997
14	0.82041	0.29118	0.10072	0.00017	0.99996
13	0.88221	0.25056	0.11371	0.00022	0.99991
12	0.95395	0.20491	0.12714	0.00030	0.99982
11	1.04771	0.15944	0.13615	0.00043	0.99969
10	1.15525	0.12596	0.14186	0.00066	0.99952
9	1.28144	0.11612	0.13763	0.00105	0.99946
8	1.44212	0.12053	0.13587	0.00169	0.99937
7	1.65172	0.13272	0.14278	0.00266	0.99911
6	1.93405	0.15273	0.15961	0.00396	0.99872
5.5	2.11595	0.16535	0.17112	0.00471	0.99850
5	2.33470	0.17688	0.18439	0.00545	0.99827
4.5	2.59873	0.19113	0.19574	0.00615	0.99814
4	2.92940	0.20890	0.20472	0.00675	0.99805
3.5	3.35070	0.23031	0.21098	0.00720	0.99798
3	3.90719	0.26058	0.21324	0.00756	0.99796
2.8	4.18384	0.27999	0.21247	0.00770	0.99800
2.4	4.88459	0.31994	0.21220	0.00792	0.99801
2	5.83351	0.37376	0.21157	0.00811	0.99798

Table S3. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **^{163}Dy** at 0 Oe in the temperature range 2-15 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R²
15	0.80669	0.35287	0.10102	0.00010	0.99997
14	0.86240	0.32644	0.10376	0.00014	0.99995
13	0.92650	0.27661	0.13364	0.00016	0.99995
12	1.00140	0.22560	0.14916	0.00020	0.99989
11	1.09990	0.20614	0.15542	0.00027	0.99986
10	1.20898	0.18310	0.16554	0.00036	0.99982
9	1.34358	0.18310	0.17573	0.00048	0.99975
8	1.51242	0.19077	0.19139	0.00060	0.99965
7	1.72893	0.21156	0.20908	0.00072	0.99959
6	2.01690	0.22844	0.23185	0.00080	0.99955
5.5	2.20004	0.24306	0.24020	0.00084	0.99954
5	2.41820	0.25295	0.24938	0.00086	0.99958
4.5	2.67923	0.30046	0.24777	0.00090	0.99955
4	3.01828	0.29785	0.26169	0.00090	0.99963
3.5	3.44736	0.32953	0.26560	0.00092	0.99964
3	4.01898	0.36854	0.27018	0.00093	0.99965
2.8	4.30593	0.38356	0.27225	0.00094	0.99965
2.4	5.02994	0.44492	0.27339	0.00096	0.99966
2	6.01177	0.51840	0.27485	0.00097	0.99967

Table S4. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **^{162}Dy** at 1000 Oe in the temperature range 4.5-15 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R²
15	0.76719	0.32071	0.12145	0.00019	0.99995
14	0.82236	0.27867	0.14585	0.00031	0.99983
13	0.88586	0.23913	0.16845	0.00050	0.99957
12	0.96188	0.18809	0.20113	0.00079	0.99904
11	1.06264	0.12504	0.23161	0.00131	0.99835
10	1.17644	0.07533	0.24780	0.00221	0.99823
9	1.31353	0.04491	0.25163	0.00406	0.99853
8	1.48767	0.03073	0.25326	0.00813	0.99893
7	1.71732	0.02299	0.25832	0.01788	0.99935
6	2.01831	0.02277	0.26492	0.04450	0.99954
5.5	2.17559	0.02730	0.26109	0.07280	0.99942
5	2.31044	0.03361	0.25035	0.11947	0.99938
4.5	2.40781	0.04207	0.23336	0.19762	0.99953

Table S5. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **^{163}Dy** at 1000 Oe in the temperature range 2-15 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R²
15	0.80654	0.34223	0.12206	0.00019	0.99993
14	0.86428	0.31084	0.14007	0.00032	0.99986
13	0.93138	0.26156	0.16920	0.00050	0.99955
12	1.01084	0.20786	0.20423	0.00077	0.99906
11	1.11718	0.14261	0.23420	0.00127	0.98380
10	1.23610	0.09500	0.24952	0.00215	0.99830
9	1.37988	0.06937	0.25254	0.00399	0.99851
8	1.56291	0.05649	0.25437	0.00792	0.99903
7	1.80577	0.05423	0.26088	0.01743	0.99939
6	2.12412	0.05833	0.26931	0.04324	0.99964
5.5	2.30272	0.06653	0.26968	0.07127	0.99962
5	2.46570	0.07743	0.26335	0.11832	0.99954
4.5	2.63172	0.09178	0.25527	0.20356	0.99956

Table S6. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **^{162}Dy@Y** at 0 Oe in the temperature range 2-15 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R²
15	0.04344	0.01002	0.21240	0.00012	0.99930
14	0.04652	0.01166	0.20149	0.00022	0.99915
13	0.05006	0.01167	0.19627	0.00039	0.99906
12	0.05437	0.00788	0.24203	0.00053	0.99841
11	0.05993	0.00547	0.25397	0.00086	0.99858
10	0.06618	0.00420	0.25272	0.00144	0.99864
9	0.07385	0.00313	0.25730	0.00254	0.99850
8	0.08345	0.00306	0.25115	0.00488	0.99912
7	0.09634	0.00305	0.26149	0.00988	0.99935
6	0.10950	0.00447	0.24614	0.02090	0.99930
5.5	0.12013	0.00469	0.25991	0.03292	0.99920
5	0.13318	0.00550	0.27632	0.05523	0.99928
4.5	0.14995	0.00613	0.30435	0.09874	0.99929

Table S7. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **^{163}Dy@Y** at 0 Oe in the temperature range 2-15 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R²
15	0.05009	0.01744	0.17803	0.00012	0.99966
14	0.05391	0.01388	0.21544	0.00018	0.99933
13	0.05803	0.01340	0.21551	0.00030	0.99928
12	0.06305	0.01095	0.24195	0.00045	0.99877
11	0.06958	0.00758	0.26459	0.00071	0.99825
10	0.07666	0.00632	0.26212	0.00120	0.99830
9	0.08541	0.00572	0.25452	0.00208	0.99885
8	0.09644	0.00609	0.24782	0.00380	0.99912
7	0.11065	0.00754	0.24229	0.00721	0.99892
6	0.12638	0.00841	0.23603	0.01308	0.99910
5.5	0.13818	0.00923	0.24212	0.01843	0.99917
5	0.15267	0.00988	0.25747	0.02608	0.99905
4.5	0.17087	0.01020	0.28085	0.03699	0.99828
4	0.19361	0.01174	0.30258	0.05268	0.99894
3.5	0.22354	0.01249	0.33594	0.07390	0.99898
3	0.26433	0.01266	0.37648	0.10238	0.99905
2.8	0.28480	0.01268	0.39339	0.11641	0.99908
2.6	0.30811	0.01291	0.40900	0.13214	0.99928
2.4	0.33561	0.01314	0.42576	0.15063	0.99936
2.2	0.37028	0.01290	0.44485	0.17270	0.99935
2	0.40254	0.01341	0.45805	0.19260	0.99947